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# PERFORMANCE ANALYSIS OF SEMIFLUIDIZED BED BIOFILM REACTORS WITH LIQUID PHASE OXYGEN (LPO) UTILIZATION

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## ABSTRACT

Performance Characteristics of Semifluidized bed biofilm reactors that deal with aerobic processes with liquid phase oxygen (LPO) Utilization have been simulated both mathematically as well as experimentally. Dispersed flow has been assumed in both packed section as well as fluidized section of the bioreactor. Accordingly, the reactor becomes equivalent to two plugflow dispersion reactors (PFDRs) in series, each with a different value of axial dispersion co-efficient. The values of design parameters of the bioreactor computed using the developed software package have been found to agree closely with experimental data collected on laboratory scale and pilot plant scale bioreactors, the maximum deviation being 12.5%.

## INTRODUCTION

Semifluidized bed bioreactors have manifested themselves as a veritable compromise between fixed bed and fluidized bed bioreactors. A semifluidized bed consists of an expanded bed [fully fluidized bed] whose expansion is restricted by a restraint (porous plate) at the top. The particles that reach the top restraint accumulate there forming a packed bed. The system thus consists of a well-fluidized bed at the bottom and a packed bed at the top. In spite of the fact that semifluidized bed bioreactors combine the merits of both packed bed and fluidized bed reactors and they function equivalent to a CSTR-PFR combination, their characteristics have been so far studied mainly experimentally and only empirical correlations are available for estimating their basic operating parameters such as semifluidization velocity, fractional fluid holdup, heights of packed section and that of fluidized section etc. A further bottleneck is that most of the experiments have been conducted on semifluidized beds employing air or water as the fluidizing medium (1) and little work has been reported on performance characteristics of semifluidized bed reactors, least of all bioreactors. Worst still, many of the experimental correlations reported predict conflicting and unacceptable values of operating parameters (1, 2), thereby making selection of the most reliable correlation difficult and confusing.

The chief operating parameters associated with semifluidized bed reactors are minimum semifluidization mass velocity ( $G_{0sf}$ ), maximum semifluidization mass velocity ( $G_{msf}$ ), voidage of fluidised section ( $\epsilon_f$ ) and volume ratio of packed section to fluidized section. Based on their experiments on a semifluidized bed composed of spherical particles and granules fluidized with water, Murthy and Roy(1) have developed the following correlations:

$$G_{0sf} / G_{msf} = 0.475 (D/d_p)^{-0.2} (\rho_s/\rho_f)^{0.17} (L_{sf}/L)^{0.38} \quad (1)$$

$$G_{msf} = 0.000185 (d_p)^{0.65} [\rho_f(\rho_s - \rho_f)]^{0.55} / (\mu_f)^{0.1} \quad (2)$$

where  $L$  = height of static bed, prior to fluidization

$$L_{sf} = \text{total height of semifluidised bed} = L_{pa} + L_f \quad (3)$$

Poddar and Dutt (3) Roy and Sharma (4) have proposed following correlations :

$$L_{pa} / (L_{sf} - L) = (1 - \epsilon_{pa}) / (\epsilon_f - \epsilon_{pa}) \quad (4)$$

$$(L_{sf} - L) / L_f = (G_{sf} - G_{0sf}) / (G_{msf} - G_{0sf}) \quad (5)$$

$$\text{where } G_{sf} = \text{operating semifluidization velocity} \quad (6)$$

Reliable correlations are scarce in literature for the estimation of the fractional fluid holdup in the fluidised section of semifluidized bed reactors. However, for a liquid-solid system, the fractional liquid holdup in the fluidized section could be estimated, with allowable error from Richardson and Zaki's correlation (5) which has been originally developed for liquid-fluidized beds.

LPO technology is a relatively recent addition towards the improved design of aerobic bioreactors (6, 7). The major operating cost of aerobic processes is that of the huge air compressors employed for forcing air into the bioreactor. In LPO technology, a calculated amount of hydrogen peroxide is added to the feed solution which releases nascent oxygen in solution. This nascent oxygen, since is highly reactive, meets the oxygen requirement of microbial growth and substrate conversion effectively. The operating cost of huge air compressors is completely eliminated. No doubt, care is to be exercised to avoid any excess use of  $H_2O_2$ , since this would lead to significant oxidative destruction of cell mass.

## MATHEMATICAL SIMULATION

The only study reported so far on semifluidized bed bioreactor is that of Narayanan and coworkers (8). It deals with immobilized enzyme reactor and the study has assumed true plug flow in the packed section of the reactor and hundred percent backmixing in the fluidized section. Though such an approximation does tally with the performance of quite a few industrial bioreactors, it induces an error on many other occasions. A more rigorous approach shall be to assume dispersed flow in both sections. The magnitude of axial dispersion coefficient ( $D_L$ ) shall be however different for the two sections, being higher for the fluidized section and lower for the packed section. In other words, the bioreactor is equivalent to two PFDRs (Plug Flow Dispersion Reactors) in series. PFDR-1 stands for the fluidized section and is followed by PFDR-2 (the packed section). This approach has been followed in the present study. The bioreactor considered is of biofilm type that conducts microbial fermentation process. Though aerobic microbial processes are dealt with in this study, due to the application of LPO technology, the bioreactor operates as a liquid fluidized semifluidized bed. Since there is no external supply of air or oxygen gas, it

ceases to be a three phase system, but gets reduced to a two phase (liquid-solid) system.

The first step in the simulation procedure is to specify the operating semifluidization velocity

( $G_{sf}$ ). This is achieved by solving equations (1), (2), (4), and (5) simultaneously by a trial and error (iterative) procedure as outlined below:

(1) A value of ( $L_{sf}/L$ ) is first assumed. For example, let

$$R = (L_{sf}/L) = 2.0 \quad (7)$$

The maximum and minimum semifluidisation mass velocities ( $G_{msf}, G_{osf}$ ) are computed from equations (2) and (1) after replacing  $d_p$  by  $d_{pm}$  and  $\rho_s$  by  $\rho_{sm}$ .

The operating semifluidization mass velocity ( $G_{sf}$ ) evidently lies between the minimum and maximum limits. A good choice is to take it equal to the arithmetic average of minimum and maximum values. Thus,

$$G_{sf} = (G_{msf} + G_{osf}) / 2 \quad (8)$$

The superficial semifluidization velocity ( $U_{sf}$ ) is then obtained as  $U_{sf} = G_{sf} / \rho_f$ .

The terminal free settling velocity ( $V_t$ ) of particle-biofilm aggregate is now computed using an iterative procedure. The value of  $V_t$  is first assumed. For example, let  $V_t = 0.1$  m/s.

The particle Reynolds number ( $Re_p$ ) is estimated as

$$Re_p = d_{pm} V_t \rho_f / \mu_f \quad (9)$$

$$\text{where } d_{pm} = (d_p + 2\delta) \quad (10)$$

$\delta$  = biofilm thickness (assumed more or less constant)

Corresponding to the above – computed value of particle Reynolds number, the value of drag coefficient ( $f_D$ ) is read from the standard plot of  $f_D$  versus  $Re_p$  (on log-log coordinates). This plot (2) is kept stored in the computer memory as a database.

The terminal free settling velocity is now computed as

$$V_t^2 = [(4/3)(\rho_{sm} - \rho_f)gd_{pm} / (\rho_f f_D)] \quad (11)$$

Where

$$\rho_{sm} = \text{density of particle - biofilm aggregate} = f\rho_m + (1-f)\rho_s \quad (12).$$

$$f = \text{volume fraction of biofilm in particle- biofilm aggregate} = 1 - (d_p/d_{pm})^3 \quad (13)$$

It is now checked whether the above- estimated value of  $V_t$  agrees with that assumed in step – 4. If not, computations are repeated from step – 5 with the newly computed value of  $V_t$ .

Once the value of  $V_t$  has been finalized through the iterative procedure described above, the fractional liquid holdup in the fluidized section ( $\epsilon_f$ ) is computed from Richardson and Zaki's correlation (5), after replacing  $d_p$  by  $d_{pm}$ . Equations (4) and (5) are now solved simultaneously for ( $L_{sf}/L$ ) and ( $L_{pa}/L$ ). The voidage of the packed section ( $\epsilon_{pa}$ ) is assumed constant and equal to 0.35.

If the above- computed values of  $R$  ( $= L_{sf}/L$ ) differs significantly from that assumed in step – 1, then a new value of  $R$  is assumed as given below and computations repeated from step – 2 :

$$R = [ R + (L_{sf}/L) ] / 2 \quad (14)$$

Once the operating semifluidization velocity ( $G_{sf}, U_{sf}$ ), fractional liquid holdup in fluidized section ( $\epsilon_f$ ), heights of fluidized section and packed section ( $L_f, L_{pa}$ ) are estimated as described above, we proceed to solve the performance equations for the two PFDRs (PFDR-1 and PFDR-2). The performance equation for PFDR-1 (fluidized section) is

$$-\frac{U_{sf}}{\epsilon_f} \frac{dC_S}{dz} + D_L \frac{d^2 C_S}{dz^2} = \eta (-r_{sp})(int) \quad (15)$$

$$\text{where } \eta = \text{effectiveness factor} \quad (16)$$

$$(-r_{sp})(int) = \text{intrinsic rate of reaction} \quad (17)$$

Aerobic processes that intrinsically follow Monod-type kinetics have been considered. However, since the process also employs LPO (Liquid Phase Oxygen) technology, an additional kinetic constant  $K_L$  has been required to be incorporated. The biomass concentration in the biofilm has been assumed to be more or less constant and equal to  $x_f$ . Accordingly,

$$(-r_{sp})(int) = (\mu_m/Y) C_{sp} x_f f [(1 - \epsilon_f)/\epsilon_f] \omega / [K_S + C_{sp}] \quad (18)$$

$$\text{where } \omega = C_N / (K_L + C_N) \quad (19)$$

$$K_L = \text{LPO utilization coefficient, kg / m}^3 \quad (20)$$

$$C_N = \text{hydrogen peroxide concentration in solution} \quad (21)$$

As stated earlier, the hydrogen peroxide concentration in the feed solution must be accurately controlled to avoid damages to microbial cells. Usually, a value of  $C_N = 2.0$  to  $5.0$  mM shall be sufficient. Since nascent oxygen is enormously reactive, it can meet the oxygen requirement of the process even at very low concentrations. Accordingly, an average value of  $C_N$  may be used in computations and it may be assumed to remain more or less constant during the process.

The effectiveness factor ( $\eta$ ), that accounts for resistance to substrate transfer into the biofilm, is computed based on the correlation proposed by Gottifreddi and Gonzo (9). This is reproduced below :

$$(1/\eta)^2 = (1/\eta_d)^2 + \exp[6(\Phi)^2 / \{5(1+\beta)^2\} - (1/\eta_d)^2] \quad (22)$$

where  $\eta_d = (\sqrt{2}/\Phi) [(1+\beta)/\beta] [\beta - \ln(1+\beta)]^{1/2} \quad (23)$

$$\Phi = L^* [\mu_m(app)/D_e K_s]^{1/2} \quad (24)$$

$$\beta = (C_{sp}/K_m) \quad (25)$$

$$\mu_m(app) = (\mu_m/Y) x_f f \omega [(1-\epsilon_f)/\epsilon_f] \quad (26)$$

$C_{sp}$  = substrate concentration at the surface of biofilm (at biofilm – liquid interface) (27)

$L^*$  = characteristic length = ( Volume of biofilm) / (surface area of biofilm)

$$= [(d_{pm})^3 - (d_p)^3] / (6 d_{pm}^2) \quad (28)$$

The substrate concentration in the liquid phase at (or very near to) the biofilm – liquid interface,

$C_{sp}$ , is estimated based on a substrate balance between the liquid bulk and the biofilm surface, as given below :

$$(k_L a)(C_s - C_{sp}) = \eta(-r_{sp})(int) \quad (29)$$

On substituting the expression for intrinsic rate from equation (18) into the above equation and simplifying, we get

$$C_{sp}^2 - [\eta\mu_m(app) / (k_L a) + K_s - C_s] C_{sp} - (K_s C_s) = 0 \quad (30)$$

The above quadratic equation is solved for  $C_{sp}$  by trial based on specified values of mass transfer coefficient ( $k_L$ ) and specified interfacial area ( $a$ ). A reasonable estimate of  $k_L$  is obtained from the correlation proposed by Koloini and coworkers (10). The interfacial area for mass transfer may be assumed equal to the surface area of biofilm and accordingly, the specific interfacial area ( $a$ ) may be defined as

$$a = (6 / d_{pm}) (1 - \epsilon_f) / \epsilon_f \quad (31)$$

The boundary conditions governing the system are,

$$\text{BC - 1 : At } z = 0, C_s = C_{s0} \quad (32)$$

$$\text{BC - 2 : At } z = L_f, C_s = C_{sb} \quad (33)$$

To note that  $C_{sb}$  is the substrate concentration in the fluid stream when it leaves the fluidized section and enters the packed section. The magnitude of  $C_{sb}$  is not known

at the outset. It is fixed during the solution of the performance equations as described subsequently.

The performance equation of the fluidized section, namely equation (15) coupled with equation

(29), is solved numerically based on the BCs specified above using a numerical algorithm, NUMCM, that utilizes line successive over – relaxation (SOR) method. Solution using a modified form of fourth order Runge – Kutta method has also been attempted. The SOR method was found to be more stable, though the values of over – relaxation factors are to be finalized by trial in advance.

The performance equation for the packed section ( PFDR – 2 ) is similar to that for PFDR – 1

and is given below:

$$-(U_{sf}/\epsilon_{pa})(dC_s/dz) + D_{LP}(d^2C_s/dz^2) = \eta(-r_s)(int) \quad (34)$$

where  $D_{LP}$  = axial dispersion coefficient for the packed section,  $m^2/s$

The effectiveness factor ( $\eta$ ) is that defined through equations (26) to (32). However, in the

packed section, it is assumed that resistance to substrate transfer in the fluid bulk is negligible and consequently,  $C_{sp} \approx C_s$ . Accordingly,

$$\beta = (C_s/K_s) \quad (35)$$

$$(-r_{sp})(int) = (-r_s)(int) = \mu_m(app)C_s/(K_s + C_s) \quad (36)$$

$$\text{where } \mu_m(app) = (\mu_m/Y) x_f f \omega[(1 - \epsilon_{pa})/\epsilon_{pa}] \quad (37)$$

The boundary conditions pertinent to the packed section are

$$\text{BC – 3: At } z = L_f, C_s = C_{sb} \quad (38)$$

$$\text{BC – 4: At } z = L_{sf} = L_{pa} + L_f, C_s = C_{se} \quad (39)$$

Equation (34) is also solved numerically based on the above – mentioned boundary conditions using the same numerical algorithm, NUMCM that involves line SOR method. To summarize, the performance equations for PFDR – 1, eqs. (15) and (29), are first solved using NUMCM starting from  $z = 0$  and continued until  $z = L_f$ , when  $C_s = C_{sb}$ . The performance equation for PFDR – 2, eq (34), is now solved numerically using the same algorithm starting from  $z = 0$  (when  $C_s = C_{sb}$ ) and proceeding upward until  $z = L_{pa}$  (when  $C_s = C_{se}$ ). The fractional conversion attained is then given by,  $\alpha = (C_{s0} - C_{se})/C_{s0}$ .



## EXPERIMENTAL STUDY (MATERIALS AND METHODS)

Experiments were conducted on laboratory scale semifluidized bed biofilm bioreactors with reactor diameter = 0.35 m, 0.5 m. Support particles = 2.5 mm granules made of polymer composites, silica and coke (each soaked with microbial sludge). Feed solutions are industrial effluents collected from food processing units, fertilizer units and paper and pulp industry. BOD of feed ranged from 300 to 700 mg/L. Feed flow rates ranged from 200 to 300 m<sup>3</sup>/hr.

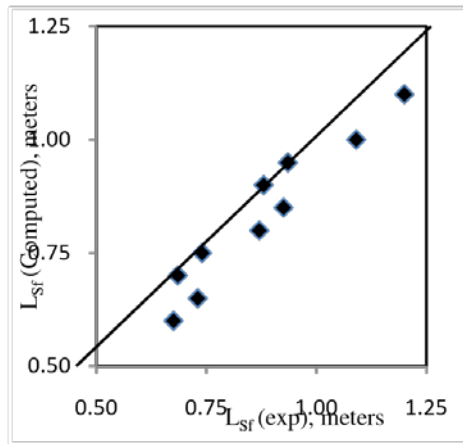
Experimental runs were also made on pilot plant scale, in two separate pilot plant reactors, the first handling waste water from a pharmaceutical industry (BOD = 350 mg/L) and the other that from a paper and pulp unit (BOD = 500 mg/L). Both reactors employed 3 mm iron oxide particles soaked with microbial sludge. Reactor diameter = 1.0 m (in both cases). Hydrogen peroxide concentration employed was 5.0 mM in all the runs. The flow rate was varied from 225 m<sup>3</sup>/hr to 325 m<sup>3</sup>/hr. Flow rates were recorded using pre-calibrated rotameters. Velometers (flow transducers) with digital display recorders were also employed to ensure accuracy. Concentrations were estimated using the conventional BOD meter as well as by using spectrophotometer and high performance liquid chromatograph (HPLC). Experimental runs were repeated at each flow rate at least thrice to establish consistency of experimental results.

## RESULTS AND DISCUSSION

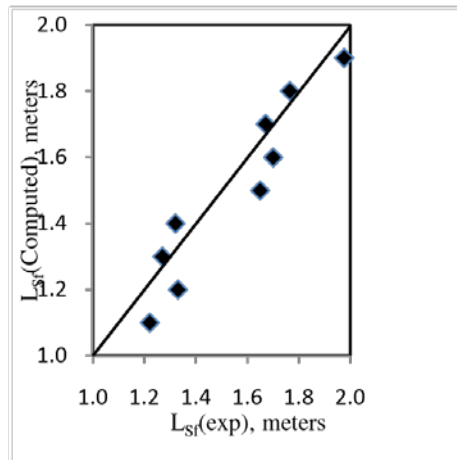
The performance characteristics of the semifluidized bed biofilm reactor computed mathematically using the developed simulation package are compared with those estimated through laboratory scale and pilot plant scale experiments. The comparison between computed values of total height ( $L_{sf}$ ) of semifluidized bed (reaction zone) and those determined experimentally is illustrated in figures (1) and (2). All the data points correspond to a fractional BOD removal (fractional conversion of substrate) of 85%, but are at different values of feed flow rate ( $Q_0$ ) and feed inlet BOD. As stated earlier, the feed flow rate ranged from 200 to 325 m<sup>3</sup>/hr and BOD level of feed solution from 300 to 700 mg/L. It can be seen from the figures that the computed results from the developed software package and the experimental results from laboratory/ pilot plant reactors agree closely with each other with a maximum deviation of 12.5%. Only a few selected sample data points are shown in figures (1) and (2), but similar agreement has been observed with all the data collected (as many as 65 data points). This ascertains the reliability of the CAD package developed.

Comparison between computed and experimental values of effluent BOD ( $C_{sf}$ ) at specific values of feed concentration ( $C_{s0}$ ), feed flow rate ( $Q_0$ ) and at given values of height of reactor zone ( $L_{sf}$ ) is shown in figures (3) and (4). In this case also, it can be seen that there is excellent agreement between the package results and experimental data, the maximum deviation being 12%. This further establishes the accuracy of the developed software package. This also ensures that the choice of the experimental correlations for estimation of the initial design parameters is satisfactorily rigorous, notwithstanding their empirical nature. The applicability of LPO ( Liquid Phase Oxygen ) technology to the operation of aerobic biofilm reactors is also herewith ascertained.

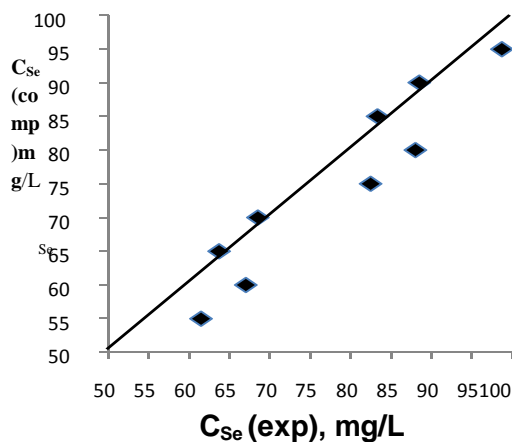




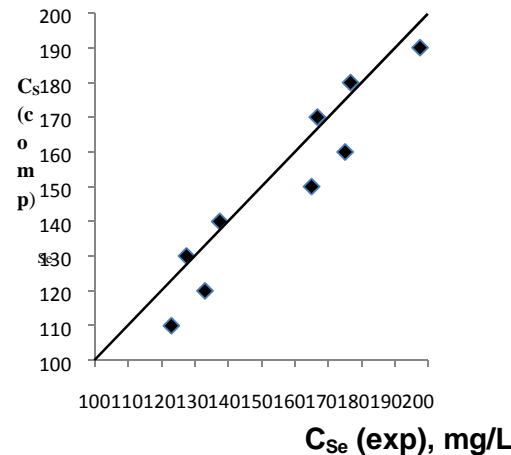
**Fig.1:** Comparison between computed & experimental values



**Fig. 2:** Comparison between computed and experimental values



**Fig. 3:** Comparison between computed and experimental values



**Fig. 4:** Comparison between computed and experimental values

## CONCLUSIONS

Performance characteristics of a semifluidized bed aerobic biofilm reactor that employs Liquid Phase Oxygen (LPO) utilization have been analyzed both mathematically as well as experimentally. A rigorous software package has been first developed through mathematical simulation of the bioreactor's performance. The package considers the reactor to be equivalent to two PFDRs (Plug Flow Dispersion Reactors) in series with different values of axial dispersion coefficient ( $D_L$ ). The performance equations were solved using a specially developed numerical algorithm NUMCM that is based on line successive over relaxation (SOR) method. No simplifying assumptions have been incorporated.

The operating features of the bioreactor estimated through the above-developed software package agree excellently with elaborate experimental data compiled on laboratory scale as well as on pilot plant scale (figures- 1,2,3,4). This demonstrates the reliability of the software package developed.

The above observation (close agreement between experimental and numerically computed results) also illustrates the applicability of the experimental correlations employed for the estimation of initial design parameters such as minimum and maximum semifluidization mass velocities fractional liquid holdup in the fluidized section of the bed and the relative size ratio of the packed section to fluidized section. No doubt, these correlations have been selected through elaborate trials.

The study also confirms the applicability of LPO (Liquid Phase Oxygen) utilization for the operation of aerobic biofilm reactors. Due to LPO utilization, the operation of the bioreactor reduces to that of a two phase system (conventional aerobic reactors being three phase systems) and it also significantly lowers the overall operating cost of the bioreactor.

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